51

**CLAIMS** 

5

1. A complex of a compound of the general formula

 $B(R^i)_3$  (1)

in which each R<sup>1</sup> independently represents an alkyl, aryl, alkylaryl, arylalkyl, cycloalkyl, alkylcycloalkyl or cycloalkylalkyl group which may be unsubstituted or substituted by one or more of the same or different substituents selected from halogen atoms and alkoxy groups;

with an organosilicon compound containing at least one primary, secondary and/or tertiary amino group.

- 15 2. A complex as claimed in claim 1, in which each R<sup>1</sup> independently represents a C<sub>1-10</sub> alkyl group, preferably an ethyl, isopropyl, *t*-butyl, *n*-butyl group.
  - 3. A complex as claimed in either claim 1 or claim 2, in which the organosilicon compound has the general formula:

20

$$\begin{bmatrix} z \end{bmatrix}_{a} \begin{bmatrix} x \end{bmatrix}_{b} \begin{bmatrix} R^{2} \\ Si \\ Q \end{bmatrix}_{c} \begin{bmatrix} L \end{bmatrix}_{d} \begin{bmatrix} X \\ Si \\ R^{2} \end{bmatrix} \begin{bmatrix} X \\ L \end{bmatrix}_{f} \begin{bmatrix} Si \\ Q \end{bmatrix}_{g} \begin{bmatrix} L \end{bmatrix}_{i} \begin{bmatrix} R^{2} \\ Si \\ R^{2} \end{bmatrix}_{k} \begin{bmatrix} X \\ P \end{bmatrix}_{g} \begin{bmatrix} R^{2} \\ R^{2} \end{bmatrix}_{c}$$

**(II)** 

25 in which:

a, q, are independently equal to 0 or 1;

b, c, d, e, f, g, i, k, p, are independently equal to or higher than 0;

(a, c, e, g, k, cannot be all equal to 0 at the same time. At least one of them should be higher than 0 and at least one of the b, d, f, i, p should be equal to or higher than 1).

each R<sup>2</sup> independently represents a hydrogen atom or a hydroxyl group or an alkyl (e.g. isopropyl, isobutyl, isooctyl, propylisobutyl, etc.), fluoroalkyl, glycidyl alkyl, acrylalkyl, (meth)acrylalkyl,

WO 2005/044867

alkoxy, alkoxyalkyl, alkenyl, cycloalkyl (e.g. cyclohexyl, propylcyclohexyl, etc.), aryl, alkyloxyaryl, aryloxyalkyl or alkyloxycycloalkyl group, each of which may be optionally substituted by one or more primary, secondary or tertiary amino groups and/or other functional groups such as hydroxyls, carbonyls; and

5 each X (which can be monovalent or divalent depending on the values of a, c, d, e, f, g, i, k, and q) independently represents a group of the general formulae (III) and (IV):

53

monovalent X

$$-R_3-N$$
 $R_5$ 

divalent X

$$R_3$$
— $R_3$ — $R_5$ 

in which R<sup>3</sup> represents an alkylene, alkenyl, phenylene or cycloalkylene group; and each of R<sup>4</sup> and R<sup>5</sup> independently represents a hydrogen atom, a hydroxyl group, or an alkyl, aryl, silylalkyl,

WO 2005/044867 PCT/EP2004/052898

54

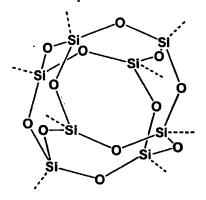
silylaryl, cycloalkyl, arylalkyl, alkylaryl, cycloalkylalkyl, alkylcycloalkyl, eterocyclic (saturated or unsaturated), phenyl (Ph-), phenoxy (Ph-O-), or Ph-(C=O)- group each of which may be optionally substituted by one or more primary, secondary or tertiary amino groups and/or other functional groups such as hydroxyls, carbonyls, etc., R<sup>4</sup> and R<sup>5</sup> can independently be also R<sup>2</sup>, R<sup>6</sup> can be a "cyclic" group that means a closed ring hydrocarbon group that is classified as an alicyclic group, aromatic group, or eterocyclic (saturated or unsaturated) group and each one of them can be mono, di-, tri-, tetra-, penta-substituted by R<sup>3</sup> or R<sup>4</sup> groups (structures IIIc, IIIe, IVc and IVe described only the mono-substituted derivatives). X can also contain organic groups or organic linking groups can include heteroatoms (e.g. O, S, Si atoms) such as in the case of heterocyclic compounds as well as functional groups (e.g. carbonyl, hydroxyl groups, etc).

 $R^7$  can be a "cyclic" of the structure  $-Si(R^2)-[Si(R^2)_2-NH-]_n-Si(R^2)$ -, where n is equal to or higher than 1.

## 15 in which L represents:

A monovalent or divalent (depending on the values of the a, b, c, e, g, k, p and q) group and can be independently selected from any of the groups representing the X group or it can also be R<sup>2</sup> or R<sup>3</sup> or R<sup>4</sup> or R<sup>6</sup> or R<sup>7</sup> or any polymeric/oligomeric organic mono- or di-radical.

## 20 in which Z represents:



where <u>every</u> silicon atom forms a bond (represented by the dashed lines on the above drawing) with
25 either X or R<sup>2</sup> or R<sup>4</sup> or R<sup>5</sup> and at least one silicon atom must be bonded with one X.

4. A complex as claimed in either claim 1 or claim 2, in which each R<sup>1</sup> is an ethyl group

WO 2005/044867 PCT/EP2004/052898

- 5. A complex as claimed in claim 3, in which the organosilicon compound has the formula derived from the general formula 11 for b=1, c=1, q=1 and a, d, e, f, g, i, k, p, q are all independently equal to 0 and where X and R<sup>2</sup> groups are as defined in claim 3.
- 6. A complex as claimed in claim 3, in which the organosilicon compound has the formula derived from the general formula 11 for b=1, c>1, e>1, k=1, p=1 and a, d, f, g, i, q are all independently equal to 0 and where X and R<sup>2</sup> groups are as defined in claim 3.
- 7. A complex as claimed in claim 3, in which the organosilicon compound has the formula 10 derived from the general formula II for a=1, b=1 and c, d, e, f, g, i, k, p, q are all independently equal to 0 and where X group is as defined in claim 3.
  - 8. A complex as claimed in claim 3, in which the organosilicon compound can be a mixture of at least two compounds as any of those claimed in claims 5 to 7.

15

- 9. A complex as claimed in claim 3, in which the organosilicon compound is 3-(2-aminoethylamino)propyltrimethoxy silane; 3-(2-aminoethylamino)propyltriethoxy silane; (aminopropyl)trimethoxy silane; (aminomethyl)trimethoxy silane; (aminomethyl)trimethoxy silane; (N-cyclohexylaminomethyl)trimethoxy silane; (N-cyclohexylaminomethyl)trimethoxy silane; (N-cyclohexylaminomethyl)trimethoxy silane; (N-cyclohexylaminomethyl)trimethoxy silane; (N-cyclohexylaminomethyl)trimethoxy silane;
- 20 cyclohexylaminomethyl)triethoxy silane; (N-phenylaminomethyl)trimethoxysilane; (N-phenylaminomethyl)methyldimethoxysilane; (N,N-dimethylaminopropyl)trimethoxysilane; Bis[(3-trimethoxysilyl)propyl]ethylenediamine; N-(3-triethoxysilylpropyl)4,5-dihydroimidazole; 2-(trimethoxysilylethyl)pyridine; Bis(p-aminophenoxy)dimethylsilane; Bis(p-aminophenoxy)dimethylsilane; Ureidopropyltrimethoxysilane;
- Bis(N-methylbenzamido)ethoxymethylsilane; Octamethylcyclotetrasilazane; 1,3-Bis(3-aminopropyl)tetramethyldisiloxane; an amino functionalised silicone fluid; an amino functionalized silica gel; an amino functionalized-POSS; or an amino/imino functionalized POSS as well as a mixture of at least two of the above.
- 30 10. A complex as claimed in claim 4, in which the organosilicon compound is hydroxyethoxysilatrane.
  - 11. The use of a complex as claimed in any one of claims 1 to 10 as an initiator for the polymerisation of a radically polymerisable monomer or oligomer.

35

12. The use of a complex as claimed in any one of claims 1 to 10 as a primer for activating a surfacein which the surface has low surface energy and comprises of polyethylene, polypropylene,

copolymers of a-olefins, fluorinated polymers (e.g polytetrafluoroethylene, etc.) and other polymers of comparable or higher surface energy in which the surface can be selected from the group of homo- or co-polymers of methyl methacrylate, polycarbonate, poly(vinyl chloride), acrylonitrile-butadiene-styrene and other plastics of comparable or higher surface energy.

5

- 13. The use of a complex as claimed in claim 12, in which the surface can be selected from the group of thermoplastics, thermosets, wood, composites, ceramics, glass, concrete, and metals.
- 14. A polymerisable composition, which comprises at least one complex as, claimed in any one
   10 of claims 1 to 10 and at least one radically polymerisable monomer and/or oligomer.
- 15. A polymerisable composition, especially a polymerisable adhesive composition, which comprises a complex as, claimed in any one of claims 1 to 10, a reactive or non-reactive diluent, at least one radically polymerisable monomer and/or oligomer, at least one decomplexing agent capable of releasing the organoborane compound from the complex upon contact and at least one radically polymerisable monomer and/or oligomer.
  - 16. A polymerizable composition as claimed in any one of the claims 14 to 15, in the form of a one-component composition.

20

- 17. A polymerisable composition as claimed in claim 14 or 15 in the form of a two-component composition in which the first part comprises at least one complex as claimed in any one of the claims 1 to 10 and a second part comprises
- at least one decomplexing agent capable of releasing the organoborane compound from the

  25 complex(es) upon contact and at least one radically polymerizable monomer and/or oligomer and/or

   an open time extender.
  - 18. A 2 part composition comprising:

Part 1: the silicon-amino organo-borane complex as claimed in any of claim 1 to 10, with optional chain extender, eg an aziridine compound;

And

Part 2: a blend of radically polymerisable compounds optionally containing toughener materials [eg ABS];

at least one decomplexer, preferably multi-functional aldehyde, and

optionally at least one open time extender, eg preferably a second radical accepting species such as 4-methylstyrene, or other alkenyl compounds.

WO 2005/044867 PCT/EP2004/052898

57

- 19. A composition as claimed in any one of the claims 13 to 18, which also contains one or more additions, preferably a natural nanoclay or a nano-organoclay, preferably atural montmorillonite or intercalated montmorillonite.
- 5 20. A composition as claimed in any one of the claims 13 to 19, which also contains trimethylolpropane tris(2-methyl-1-aziridinepropionate).
- 21. A composition as claims in any one of the claims 13 to 20, which also contains at least one material that manages the heat of polymerization reaction such that adhesion to the substrates is 10 maintained.
  - 22. A polymeric composition as claimed in any of the claims 13 to 21, in which the concentration of complex is sufficient to provide 0.001 to 10.0%w, preferably 0.002 to 7.0%w, and most preferably 0.003 to 5.0% of boron, based on the total weight of the composition.